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Determination of amplitudes and times of magnetic induction and photon echoes from the pulse matrix

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The formalism of the T-matrix for the treatment of interaction between radiation and matter is extended to the problem of magnetic induction and photon echoes. The cases of two and three pulses have been discussed. Some observations are made on the connection between the echo sequences and the time relations among the incident pulses.

1. INTRODUCTION

The dynamics of an atom can be visualised as the motion of a vector $\boldsymbol{r}$ determined by the wave function of a two-level system with eigen states ψ_a, ψ_b interacting with radiation (Feyman *et al* 1957). For an assembly of atoms labelled by $j = 1, 2, \ldots$, the expectation value of the polarisation μ is given by

$$< \boldsymbol{\mu}_{op} > = \tfrac{1}{2}\gamma \Sigma \boldsymbol{r}_j = \tfrac{1}{2}\gamma p(\boldsymbol{r})_{av}, \qquad \ldots (1)$$

where $\gamma = (\mu_x + i\mu_y)_{ab}$, p the number of two-level systems and $(\boldsymbol{r})_a$ the average value of $\boldsymbol{r}$ per system.

The phenomenon of pulse-like radiations in spontaneous radiative processes called photon echoes analogous to magnetic induction echoes observed by Hahn (1954) is essentially emission from correlated states of the atoms of an assembly which momentarily is in a super-radiant state as was pointed out by Abella *et al* (1966). The superradiance originally discussed by Dicke (1954) corresponds however to the circumstances of free magnetic induction decay observed by Hahn (1954). The problem was discussed from a more general and systematic point of view by Venkatesh & Roy (1971). Two radiation pulses are required for the production of a photon echo, the first to tip the correlated moment along the (pseudo) static field to the plane transverse to the field, and the second to drive individual moments precessing with random angular velocities in an inhomogeneous field back to the *line* where they are in step again after a definite interval of time.

The problem considered here is the response of an assembly of similar atom to two and three incident radiation pulses when the centre frequency of the radiation is equal to the transition frequency w_0 of the isolated pair of spectral levels of the atom. The response is in the form of a sequence of echo pulses bearing definite time-relations to each other within the relaxation time T_2. The treatment is applicable unifoımly to magnetic induction and photon echoes.

2. The Pulse Matrix

Let us consider a rotating coordinate system Σ' of which the 3'-axes coincides with the 3-axis of the fixed system Σ, the angular velocity of rotation being w, the frequency of the incident radiation (Venkatesh & Dixit 1970, 1971).

The matrix representing a rotation of the quasimoment in due to the action of the radiation pulse is given by

$$T = \begin{pmatrix} \cos \theta/2 - \frac{iw'_3}{\Omega} \sin \theta/2 & \frac{-iw'_1}{\Omega} \sin \theta/2 \\ -\frac{iw'_1}{\Omega} \sin \theta/2 & \cos (\theta/2) + \frac{iw'_3}{\Omega} \sin \theta/2 \end{pmatrix}, \qquad \dots (2)$$

where $w'_3 = w_0 - w$; $\Omega = (w'^2_1 + w'^2_2 + w'^2_3)^{\frac{1}{2}}$. The pulse strength is given by

$$\Omega \Delta t = \theta, \qquad \dots (3)$$

Δt being the duration of the pulse. For the transition $\Delta m = 0$ which we shall consider here for the sake of simplicity $w'_2 = 0$. Under the approximation

$$w'_3 = \delta w \approx 0. \qquad \dots (4)$$

The radiation matrix is

$$T_0 = \begin{pmatrix} \cos \theta/2 & -i \sin \theta/2 \\ -i \sin \theta/2 & \cos \theta/2 \end{pmatrix}. \qquad \dots (5)$$

Eq. (4) reflects a physical condition, namely the slight inhomogeneity of the field. The field inhomogeneity determines the relaxation time T_2 by the relation

$$T_2 = \frac{1}{\Delta w}, \qquad \dots (6)$$

where Δw is the spread of the transition frequencies, and determines the time scale for the occurrence of a coherent set of echoes. It plays the decisive role in the production of echoes.

The precessional matrix representing the free rotation of the vector $\boldsymbol{r}$ about the 3-axis is given by

$$T_\varphi = \begin{pmatrix} \exp (i\phi/2 & 0 \\ 0 & \exp(-i\phi/2) \end{pmatrix}, \qquad \dots (7)$$

where

$$\phi = w'_3 t = \delta w.t. \qquad \dots (8)$$

The whole effect of field inhomogeneity is contained in this matrix. Note $\Delta t << t$. The matrix $T_\theta T_\varphi$ which may be called the pulse matrix $T^{(k)}$ for the kth pulse irradiating an assembly of atoms is given by the unitary matrix

$$T^{(k)} = \begin{pmatrix} T_{11}^{(k)} & T_{12}^{(k)} \\ T_{21}^{(k)} & T_{22}^{(k)} \end{pmatrix} = \begin{pmatrix} \cos\frac{\theta_k}{2}\exp(i\phi_k/2) & -i\sin\frac{\theta_k}{2}\exp(i\phi_k/2) \\ -i\sin\frac{\theta_k}{2}\exp(-i\phi_k/2) & \cos\frac{\theta}{2}\exp(-i\phi_k/2) \end{pmatrix}, \qquad \dots (9)$$

ϕ_k is the angle traced by the precessing quasi-moment of an individual atom in the 1–2 plane after the kth pulse is applied at the instant t_k:

$$\phi_k = \delta w(t-t_k). \qquad \dots (10)$$

The final state of a single atom irradiated by a sequence of pulses is obtained from the transformation of the initial state of the system by the product matrix T. Let us transform the reduced density matrix $\mu = 2\rho - 1$:

$$T\mu(0)\bar{T} = T^{(N)}T^{(N-1)} \dots T^{(2)}T^{(1)}\mu(0)\bar{T}^{(1)}\bar{T}^{(2)} \dots \bar{T}^{(N-1)}\bar{T}^{(N)} = \mu_t\,, \qquad \dots (11a)$$

or

$$\mu_{(t)} = \begin{pmatrix} T_{11} & T_{12} \\ T_{21} & T_{22} \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \begin{pmatrix} T_{11} & T_{12} \\ T_{21} & T_{22} \end{pmatrix} = \begin{pmatrix} r_3 & r_1 - ir_2 \\ r_1 + ir_2 & -r_3 \end{pmatrix} \qquad \dots (11b)$$

The unitary matrices $T^{(k)}$ and the product matrix T have the general structure

$$T^{(k)} = \begin{pmatrix} A_k & B_k \\ -\bar{B}_k & \bar{A}_k \end{pmatrix}, \quad T = \begin{pmatrix} A & B \\ -\bar{B} & \bar{A} \end{pmatrix}, \qquad \dots (12a)$$

where

$$A_k\bar{A}_k + B_k\bar{B}_k = 1; \quad A\bar{A} + B\bar{B} = 1 \qquad \dots (12b)$$

Thus r_1, r_2 and r_3 and hence the relevant physical quantities are obtained directly by the elements of the product matrix, T_{11} and T_{12}.

$$r_1 - ir_2 = -2T_{11}T_{12} = -2AB \qquad \dots (13)$$

$$r_3 = |T_{11}|^2 - |T_{12}|^2 = A\bar{A} - B\bar{B}. \qquad \dots (14)$$

3. Two and Three Radiation Pulses

The special cases of two and three pulses can be treated simply by using the unitary property of T in eq. (12) although the case of four pulses already involves very heavy work by this method.

We have from eqs. (9) and (12)

$$A_k\bar{A}_k = \tfrac{1}{2}(1+\cos\theta_k), \qquad \bar{A}_kB_k = -\tfrac{1}{2}i\sin\theta_k,$$
$$B_k\bar{B}_k = \tfrac{1}{2}(1-\cos\theta_k), \qquad A_k\bar{B}_k = \tfrac{1}{2}i\sin\theta_k,$$
$$A_k\bar{A}_k-B_k\bar{B}_k = \cos\theta_k, \qquad \bar{A}_kB_k-A_k\bar{B}_k = -i\sin\partial_k. \tag{15a}$$

Also,

$$A_k^2 = A_k\bar{A}_k\exp(i\phi_k) = \tfrac{1}{2}(1+\cos\theta_k)\exp(i\phi_k),$$
$$B_k^2 = -B_k\bar{B}_k\exp(i\phi_k) = \tfrac{1}{2}(1-\cos\theta_k)\exp(i\phi_k),$$
$$A_kB_k = \bar{A}_kB_k\exp(i\phi_k) = -\tfrac{1}{2}i\sin\theta_k\exp(i\phi_k),$$
$$A_k^2-B_k^2 = \exp(i\phi_k). \tag{15b}$$

From eq. (13) we have for two incident pulses with a time interval of τ secs.

$$r_1-ir_2 = -2[A_2\bar{A}_2\bar{A}_1B_1\exp i(\phi_2+\phi_1)+B_2\bar{B}_2A_1\bar{B}_1\exp i(\phi_2-\phi_1)$$
$$+\bar{A}_2B_2(A_1\bar{A}_1-B_1\bar{B}_1)\exp(i\phi_2)]. \quad \ldots \tag{16}$$

From eq. (1) the polarisation of the assembly is obtained by taking the appropriate distribution function for the random variable δw and integrating over the width Δw. Note $\phi_k = \delta w.t_k$. Let us take the Gaussian distribution function

$$P(\delta w) = \frac{T_2}{(2\pi)^{\frac{1}{2}}}\exp[-\tfrac{1}{2}(\delta wT_2)^2]. \tag{17}$$

The average polarisation per atom is then given by integrating the expression (16) over δw (or ϕ)

$$\tfrac{1}{2}(r_1-ir_2)_{av} = \int_{\Delta w}(r_1-ir_2)P(\delta w)d\delta w.$$

The sole contribution to the echo amplitude is made by the middle term of eq. (16) after integration

$$(r_2)_{av} = \tfrac{1}{2}(1-\cos\theta_2)\sin\theta_1h(2\tau), \tag{18}$$

where

$$h(t') = h(\Delta w, t, t') = \exp[-(t-t')^2/2T_2^2]. \quad \ldots \tag{19}$$

Note when $t = t' = 2\tau$, the intensity is maximum corresponding to an echo. The total polarisation is found by multiplying eq. (18) by the total number of two-level systems p in the assembly so that the internsity of the emitted radiation determined by $\tfrac{1}{2}\gamma p(r_2)_{av}$ is proportional to p^2.

For three pulses incident at time 0, τ and T

$$T_{11}T_{12} = AB = \{A_3(A_2A_1-B_2\bar{B}_1)-B_3(\bar{B}_2A_1+A_2\bar{B}_1)\}$$
$$\times\{A_3(A_2B_1+B_2\bar{A}_1)+B_3(-\bar{B}_2B_1+\bar{A}_2\bar{A}_1)\}, \tag{20}$$

on expansion

$$\begin{aligned}
\tfrac{1}{2}(r_1-ir_2) = -AB = {} & \bar{A}_3B_3(A_2\bar{A}_2-B_2\bar{B}_2)(A_1\bar{A}_1-B_1\bar{B}_1)\exp(i\phi_3) \\
& +2\bar{A}_3B_3\bar{A}_2B_2A_1\bar{B}_1\exp i(\phi_3-\phi_1))+2\bar{A}_3B_3A_2\bar{B}_2\bar{A}_1B_1\exp(i(\phi_3+\phi_1)) \\
& +A_3\bar{A}_3\bar{A}_2B_2(B_1\bar{B}_1-A_1\bar{A}_1)\exp i(\phi_3+\phi_2))+ \\
& +B_3\bar{B}_3A_2\bar{B}_2(A_1\bar{A}_1-B_1\bar{B}_1)\exp(i(\phi_3-\phi_2))+ \\
& +A_3\bar{A}_3B_2\bar{B}_2A_1\bar{B}_1\exp(i(\phi_3+\phi_2-\phi_1))+ \\
& +B_3\bar{B}_3A_2\bar{A}_2\bar{B}_1A_1\exp(i(\phi_3-\phi_2-\phi_1))- \\
& -B_3\bar{B}_3B_2\bar{B}_2\bar{A}_1B_1\exp(i(\phi_3-\phi_2+\phi_1))- \\
& -A_3\bar{A}_3A_2\bar{A}_2\bar{A}_1B_1\exp(-i(\phi_3+\phi_2+\phi_1)). \qquad \ldots (21)
\end{aligned}$$

Integration over δw after substitution from eq. (15) leads to the expression for $(r_2)_{av}$ and hence to the expectation value of polarisation (1) :

$$\begin{aligned}
(r_2)_{av} = {} & \tfrac{1}{4}(1-\cos\theta_3)(1+\cos\theta_2)\sin\theta_1 h(2T) \\
& +\tfrac{1}{2}(1-\cos\theta_3)\sin\theta_2\cos\theta_1 h(2T-\tau) \\
& -\tfrac{1}{4}(1-\cos\theta_3)(1-\cos\theta_2)\sin\theta_1 h(2T-2\tau) \\
& +\tfrac{1}{2}\sin\theta_3\sin\theta_2\sin\theta_1 h(T+\tau) \\
& +\tfrac{1}{4}(1+\cos\theta_3)(1-\cos\theta_2)\sin\theta_1 h(2\tau). \qquad \ldots (22)
\end{aligned}$$

With time the successive terms of eq. (22) became dominant when the condition $t = t'$ in eq. (19) is satisfied. These large transient polarisations give rise to the sequence of echoes. In the following table the echoes are labelled and the time of the echoes are indicated.

$A_2 : 2\tau$	$p_2 : \tau$	$p_1 : 0$
$A_3 : 2T$	$B_3 : 2T-\tau$	$a_3 : 2T-2\tau (T > 2\tau)$
$C_3 : T+\tau$	$p_3 : T$	$x_3^+ : T-\tau.$

There are thus five echo pulses appearing at the time $2T$, $2T-\tau$, $2T-2\tau$, $T-\tau$, 2τ. p_1, p_2, p_3 denote the incident pulses. The echo a_3 is a secondary (conditional) echo and x_3^* does not occur at all (see the following section 4).

4. Echo Sequences

Two sets of echoes can be distinguished. The Primary echoes imply no conditions on time other than the given conditions :

$$T > \tau > 0. \qquad \ldots (23)$$

In addition there are secondary echoes which imply conditions on the times of the incident pulses for their occurrence. For example there is one secondary echo in the case of three incident pulses, namely that occurring at $2T-2\tau$ for this should satisfy

$$2T-2\tau > T \quad \text{or} \quad T > 2\tau. \qquad \ldots (24)$$

The condition expresses the causal principle that an echo can occur at $2T-2\tau$ after the third incident pulse has interacted with the system at T.

In any observed system of echoes distinct sequences of echoes can be identified. The special time relations (apart from eq. (23)) determine not only the occurrence of secondary echoes but also the matching of the sequences or interposition of one sequence into another. For instance if all the possible echoes were to appear condition (24) must be satisfied and the longest sequences could be formed with further conditions.

$$2T > 2T-\tau > 2T-2\tau > T+\tau > 2\tau\,, \qquad \text{if } T > 3\tau$$

$$2T > 2T-\tau > T+\tau > 2T-2\tau > 2\tau\,, \qquad \text{if } 2\tau < T < 3\tau$$

The sequences are

$$A_3\, B_3\, a_3\, C_3\, A_2\,, \qquad \text{if } T > 3\tau$$

$$A_3\, B_3\, C_3\, a_3\, A_2\,, \qquad \text{if } 2\tau < T < 3\tau$$

5. Summary and Discussinn

The method of treating the problem of interaction of radiation with matter with the help of the rotation matrix eq. (9) previously developed for dealing with problems of emission from correlated states and from maser-like devices is here extended to the problem of photon and magnetic induction echoes. The dependence of the number and sequence of echoes on the time relations has also been pointed out. This additional feature becomes extremely interesting from the point of view of applications (e.g., code, holophony) if one could deal with a large number of incident pulses. The direct use of the unitary property of T in the way we have shown here would however be powerless to deal with the general problem but the rotation matrix provides the one elegant method of dealing with it.

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